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MASTER review

TENSILE CREEP PERFORMANCE OF A DEVELOPMENTAL, IN-SITU REINFORCED SILICON NITRIDE¹

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ABSTRACT

The creep performance of a developmental, in-situ reinforced silicon nitride was evaluated at temperatures between 1300-1425°C in ambient air. The minimum creep rate as a function of tensile stress and temperature was evaluated, and the measured tensile creep performances of two different specimen geometries (buttonhead and dogbone - machined from same billet of material) were compared. This silicon nitride exhibited comparable, or better, creep resistance than other silicon nitrides described in the literature. The measured creep response of the material and lifetime were observed to be geometry dependent; the smaller cross-sectioned dogbone specimens exhibited faster creep rates and shorter lives, presumably due to faster oxidation-induced damage in this geometry. The tensile creep rates and lifetimes were found to be well-represented by the Monkman-Grant relationship between 1350 and 1425°C, with some evidence suggesting stratification of the data for the 1300°C tests and a change in dominant failure mode between 1300 and 1350°C. Lastly, values of the temperature-compensated stress exponent and activation energy for tensile creep were found to decrease by approximately 80 and 75% in compression, respectively, illustrating anisotropic creep behavior in this silicon nitride.

I. INTRODUCTION

Silicon nitride (Si_3N_4) remains a leading candidate material for use in structural components (e.g., nozzles and blades [1]) in advanced gas turbine engines. Si_3N_4 offers greater operating temperatures than currently used superalloys, is less dense which results in less inertial effects on performance, and offers the possibility of component design without the necessary provisions for cooling. In addition, Si_3N_4 has good thermal shock resistance and high thermal conductivity, is relatively corrosion-resistant, and possesses good high temperature strength.

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should this be the Heavy Vehicle Production System Materials Program

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For the present study, interest existed to mechanically characterize the high temperature deformation behavior of a developmental Si_3N_4 that is a candidate material for gas turbine engine components. Examined test temperatures bracketed 1370°C (2500°F), as 1370°C has been cited as a desirable turbine inlet temperature which will produce more efficient and powerful gas turbine engines with less NO_x emissions [2]. The high temperature tensile creep performance was examined as a function of test specimen geometry (i.e., test volume), and compared to both its compressive creep performance and the creep resistance of other candidate Si_3N_4 materials. lower ←

II. EXPERIMENTAL PROCEDURES

The laboratory grade of the in-situ reinforced silicon nitride, designated as SN5-L,² was sintered above 1900°C under elevated nitrogen pressures to attain >99% theoretical density. The SN5-L contained approximately 10 wt% of proprietary rare earth oxides as sintering aids. The *in-situ* reinforcement was induced through the preferential growth of acicular-shaped $\beta\text{-Si}_3\text{N}_4$ grains, which formed an interlocking network of grains. The microstructure of SN5-L is illustrated in Fig. 1. The acicular grains are mixed with sub-micron equiaxed grains. A thorough description of the material can be found in Ref. [3].

Uniaxial tensile creep tests were conducted at temperatures between 1300-1425°C within a stress range of 75-300 MPa. A buttonhead specimen geometry (cylindrical gage section: 6.35 mm dia. x 35 mm length) was employed for the majority of the tests; however, tensile creep tests were also performed at 1400°C using a dogbone specimen geometry (prismatic gage section: 2.54 mm x 2.54 mm x 25.4 mm length). Dogbone specimens were machined from the same billets as the buttonhead specimens, so that the starting material for both specimen types were believed to be the same. The two tensile specimen geometries are shown in Fig. 2. Contact extensometry was employed for the creep tests conducted with the buttonhead specimen, while laser extensometry was employed with the dogbone specimen; both specimen geometries, test systems, and procedures have been described in the literature [4-5]. Tensile creep strain was measured as a function of time and monitored via computer-aided data acquisition during all tests. was ←

Fig. 1. Polished and plasma-etched microstructure of SN5-L Si_3N_4 .

2 The "SN5-L" composition is a 1995-vintage laboratory-grade silicon nitride processed in an experimental furnace at AlliedSignal, Morristown, NJ.

Fig. 2. The relative sizes of the buttonhead and dogbone tensile specimens.

Uniaxial compression creep tests were also conducted as a supplement to examine any trends in creep anisotropy. A cylindrical-shaped specimen having a diameter of 5.00 mm and a length of 10.00 mm was used. Aligned α -SiC pushrods were used to load the specimen ends, and the same style of contacting extensometer used in the tensile creep tests was used to monitor the compressive creep strain. The compressive creep strain was measured as a function of time and monitored via computer-aided data acquisition.

For the post-testing creep analysis, the steady-state or minimum creep rate was examined as a function of stress (tension and compression) and temperature after the power law relation, $\{\dot{\epsilon} = A\sigma^n \exp(-Q/RT)\}$, where $\dot{\epsilon}$ is the minimum creep rate, A is a pre-exponential constant, σ is the applied stress, n is the creep rate - stress exponent, Q is the activation energy, R is the gas constant, and T is absolute temperature. Creep rate - stress exponents and activation energies were determined using multi-linear regression of the tension and compression creep data (after the natural logarithm of both sides of the power law relation were taken) and interpreted. The applicability of the Monkman-Grant relation [6], $\{t_f = C(\dot{\epsilon})^{-m}\}$, where t_f is the time to failure, and C and m are constants, was also examined.

III. RESULTS & DISCUSSION

IIIA. Tensile Creep Performance of SN5-L

The minimum tensile creep rate as a function of applied tensile stress is illustrated in Fig. 3 for the tests conducted at 1300, 1350, 1370, 1400, and 1425°C. Creep data for the whole test matrix is listed in Table 1. The number of buttonhead specimens was limited, so only a minimum of two creep tests per temperature were permissible. The amount of data is insufficient for quantifiable characterization of creep behavior, but qualitative trends were sought nonetheless in the present study. The creep rate - stress exponents between 1350-1400°C are comparable ($\approx 3-4$ in value), suggesting that a similar dominant creep mechanism was active throughout this temperature range. This value in creep rate - stress exponent is consistent with an exponent value for another silicon nitride [7] where the authors of that study attributed this n value to cavitation accompanied by grain separation by viscous flow, and perhaps grain boundary sliding. The presence of two- and multi-grain cavities was indeed identified (see Fig. 4) during fractography with the scanning electron microscope. The presence of two-grain cavities in a similar grade of SN5-L was previously attributed to grain-grain contact and the dislocation-nucleation of cavities [3]. However, the size of two-grain cavities ($\approx 50-100$ nm) tended to be substantially less than multi-grain junction cavities ($\approx 100-500$ nm) in SN5-L, so the effects and cumulative volume of multi-grain junction cavities likely

accounted for the majority of the total measured tensile creep strain. Multilinear regression yielded $n \approx 1$ at 1425°C, but the variability in the three datapoints for this temperature shown in Fig. 3 may account for n being perhaps misleadingly low-valued. For example, $n \approx 3$ for 1425°C if the 100 MPa datapoint were omitted; this value would indicate that the dominant creep mechanism that was operative at 1425°C was the same as that between 1350-1400°C. The creep rate - stress exponent was different at 1300°C, $n \approx 13$, suggesting that another dominant mechanism of creep ~~were~~ responsible for the exhibited behavior at this temperature. The fracture surfaces of specimens tested at 1300°C showed definitive mirror, mist, and hackle markings, while tests at temperatures ~~at~~ 1350°C resulted in creep-damage-induced or stress-oxidation-induced failures [4]. From this, it may be concluded that there was a change in dominant failure mode from slow-crack-growth-assisted failure to creep-damage-accumulation-assisted failure between 1300 and 1350°C for SN5-L.

The measured creep performance of SN5-L appeared to be a function of specimen geometry. The smaller-cross-sectioned dogbone specimen, see Fig. 2, exhibited faster creep rates than the buttonhead specimen, as illustrated by the 1400°C data in Fig. 5. The surfaces of the crept SN5-L specimens were heavily oxidized (i.e., an oxide scale was visible to the naked eye) as a result of testing at 1370°C and above. Two facts may be first stated whose combination may result in the observed faster creep rates of the dogbone specimen: the dogbone cross-sectional area is much smaller ($2.5 \times 2.5 \text{ mm} = 6.25 \text{ mm}^2$) than that of the buttonhead specimen ($6.35 \text{ mm dia.} = 31.67 \text{ mm}^2$); cations that are necessary as reactive species for the continued growth of the oxide scale do not have as far to diffuse in the dogbone specimen as they do in the buttonhead specimen. With the mass transport of the

Fig. 3. Minimum tensile creep rate as a function of applied tensile stress. Buttonhead specimens used to generate this data.

metal cations in the secondary phase to the surface likely creating pores and voids [4, 8-9] in the near-surface volume of both specimen geometries, a situation is created during the creep damage accumulation of both SN5-L specimen types where (1) the net compliance increase of the SN5-L dogbone specimen is greater than that for the SN5-L buttonhead specimen (at identical test conditions), and (2) the dogbone specimen exhibits ~~faster~~ creep rates and shorter lifetimes as a consequence. The creep-rate stress exponents were equivalent for both specimen geometries though, indicating that the activity of the dominant creep mechanism was independent of specimen geometry.

Fig. 4. Both the smaller two-grain (TG) and larger multi-grain (MG) cavities were present in crept SN5-L (Microstructure of specimen 3-DB shown).

Fig. 5. Minimum tensile creep rate as a function of tensile stress, temperature, and specimen geometry. At 1400°C, the dogbone specimen geometry resulted in faster creep rates than the buttonhead specimen geometry.

When the creep rate of SN5-L was graphed as a function of time to failure (i.e., the Monkman-Grant relation) in Fig. 6, a power law fit of the data showed $m = 1$, indicating that the accumulation of creep damage is the fatigue-limiting failure mechanism in SN5-L over the temperature and stress regimes examined. It may be argued that the two 1300°C data points in Fig. 6 are stratified from the rest of the data; this would be consistent with the higher n value and the slow-crack-growth failures observed during fractography of the original fracture surfaces. This stratification is another indication that there is a change in the dominant failure mode in SN5-L between 1300 and 1350°C.

Fig. 6. With $m \approx 1$ in the Monkman-Grant relation, it is evident that the creep damage mechanism serves as the same lifetime-limiting mechanism. However, the apparent stratification of the 1300°C data suggests that a different dominant mechanism exists at temperatures 1350°C and above.

IIIB. Comparison of Tensile Creep Performance

SN5-L appears to exhibit comparable, or better, creep resistance than SN88³ [10-11] and NT154⁴ [12] silicon nitrides. Creep data generated with buttonhead specimens for all three silicon nitrides is shown in Fig. 7, and the limited data for SN5-L shows that it is more creep resistant than NT154 and comparable with SN88.

³ Manufactured by NGK Insulators, LTD., Nagoya, Japan.

⁴ 1992 vintage, manufactured by Norton Advanced Ceramics, Northboro, MA.

Fig. 7. The creep resistance of SN5-L is comparable, or better, than that for SN88 or NT154 silicon nitrides.

IIIC. SN5-L Creep Asymmetry

SN5-L exhibited greater creep resistance in compression than in tension. An example of this anisotropy is illustrated in the tension and compression creep history examples in Fig. 8. Tension tests at 1400 and 1425°C exhibited a tertiary creep regime, while those below 1400°C and those in compression did not. The creep rate in compression was approximately an order of magnitude lower in compression than in tension at 1425°C for the same magnitude of stress. This anisotropy is consistent with other structural monolithic ceramics described in the literature [13-15]. Because of the creep rate anisotropy, the calculated creep rate - stress exponent and activation energies also exhibited anisotropy as a consequence, as indicated in Fig. 9. The creep rate - stress exponent and activation energy decreased from $n_t = 4.0$ and $Q_t = 1080$ kJ/mol in tension to $n_c = 0.8$ and $Q_c = 260$ kJ/mol in compression (a drop of approximately 80% and 75% for n and Q , respectively). The higher activation energy for tensile creep is consistent with viscous deformation of a yttrium oxynitride secondary phase [16], while the lower-valued activation energy for compressive creep is consistent with atomic diffusion through the secondary phase [17]. The decreases in creep rate - stress exponent and activation energy are indicative of a difference in dominant creep mechanism of cavitation in tensile creep and diffusion in compressive creep. This anisotropy is important for it requires that modelers of creep be cognizant of the sign of stress throughout in their creeping component, and the incongruent creep behavior (and fatigue) that will be a consequence.

What do you really mean here? What is a "creeping component"? a bug?

I thought you said that the rare-earth oxide additives were "proprietary" earlier.

Fig. 8. Comparison of creep histories of SN5-L tested in tension or compression.

Fig. 9. The creep anisotropy of SN5-L is evidenced by the differences in its creep rate - stress exponent and activation energies in tension and compression.

IV. CONCLUSIONS

SN5-L silicon nitride exhibited comparable, or better, creep resistance than SN88 or NT154 silicon nitrides. The measured creep response and lifetime of the SN5-L were observed to be geometry dependent; the smaller-cross-sectioned dogbone specimens exhibited faster creep rates and shorter lives than buttonhead specimens due to more rapid oxidation. An equivalence in creep rate - stress exponent and fatigue - stress exponent (i.e., $m \approx 1$ for the Monkman-Grant relation) suggests that the accumulation of creep damage serves as the fatigue- or lifetime-limiting mechanism. Evidence suggests there is stratification of the creep rate -lifetime data for the 1300°C that is indicative of a change in the dominant failure mode between 1300 and 1350°C for SN5-L. The values of the temperature-compensated stress exponent and activation energy for tensile creep were found to decrease by approximately 80 and 75% for compressive creep, respectively, illustrating strong creep-anisotropic behavior in this silicon nitride. The higher valued creep rate - stress exponent and activation energy in tension are indicative of cavitation-associated creep, while the lower valued exponent and activation energy in compression are characteristic of diffusion-associated creep.

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Table. 1. Test Matrix and Creep Results.

Specimen ID	Temp. (°C)	Stress (MPa)	Minimum Creep Rate - ($\times 10^{-10}/s$)	Time to Failure (h)
AS544A-BH	1300	250	21	208
AS544B-BH	1300	300	240	35.7
AS540B-BH	1350	100	6.3	2686*
AS540A-BH	1350	200	58	249
AS8004-BH	1370	75	3.4	3200*
AS5411B-BH	1370	100	10	
AS541A-BH	1400	75	11	1732*
AS5412B-BH	1400	100	43	
AS8001-BH	1425	50	94	381
AS8003-BH	1425	75	260	140
AS8002-BH	1425	100	180	175
1-DB	1400	75	47	680.3
2-DB	1400	100	490	43.9
3-DB	1400	100	450	88.6
4-DB	1400	150	1800	9.1
5-DB	1400	150	1200	30.2
6-DB	1400	150	900	26.0
7-DB	1400	150	2200	13.9
AS411-C	1300	-100	3.7	*
AS540B21-C	1300	-300	9.7	*
AS540B11-C	1370	-100	9.9	*
AS422-C	1370	-200	15	*
AS421-C	1425	-100	16	*

BH = Buttonhead tensile specimen

DB = Dogbone tensile specimen

C = Cylindrical compression specimen

* = Test interrupted (no failure)